

We would like to thank the reviewer for useful comments. In the following we answer the specific comments (included in “**boldface**” for clarity) and, whenever required, we describe the related changes implemented in the revised manuscript.

Anonymous Referee #3

Overview

The paper presents the results of an analysis of the new MIPAS CCl₄ product from the ESA processor. While opportunities for validation are limited, the authors do exploit one of the strengths of MIPAS, which is a 10-year globally sampled dataset to draw conclusions on interhemispheric variation and trends. On the whole, the paper is a clearly-written and convincing and I have no major criticisms.

General comments

a) While there is a convincing trend (matching the ground stations) it would have been useful to apply the same trend analysis to a different molecule retrieved with the same algorithm (eg N₂O?) which has no expected trend. This would help quantify the contribution of any calibration drift.

We have repeated the trend analysis for the N₂O and, at least for pressures between 60 and 200 hPa we do not find any statistically significant trend at all latitudes. This finding confirms that the residual calibration drift error of MIPAS is very small, as already anticipated by the careful Level 1b studies carried-out by the MIPAS Quality Working Group team and already cited in the paper (see Sect. 2.1). In the revised paper, still we are not showing maps of N₂O trends which are not a focus of the current study, we are considering N₂O trends for a future additional publication.

b) Of all the time-series fit parameters, it would have been helpful to indicate which ones were actually significant: the trend, constant and annual cycles are obvious from Fig 10 but what effect do the other terms have? Were they really needed?

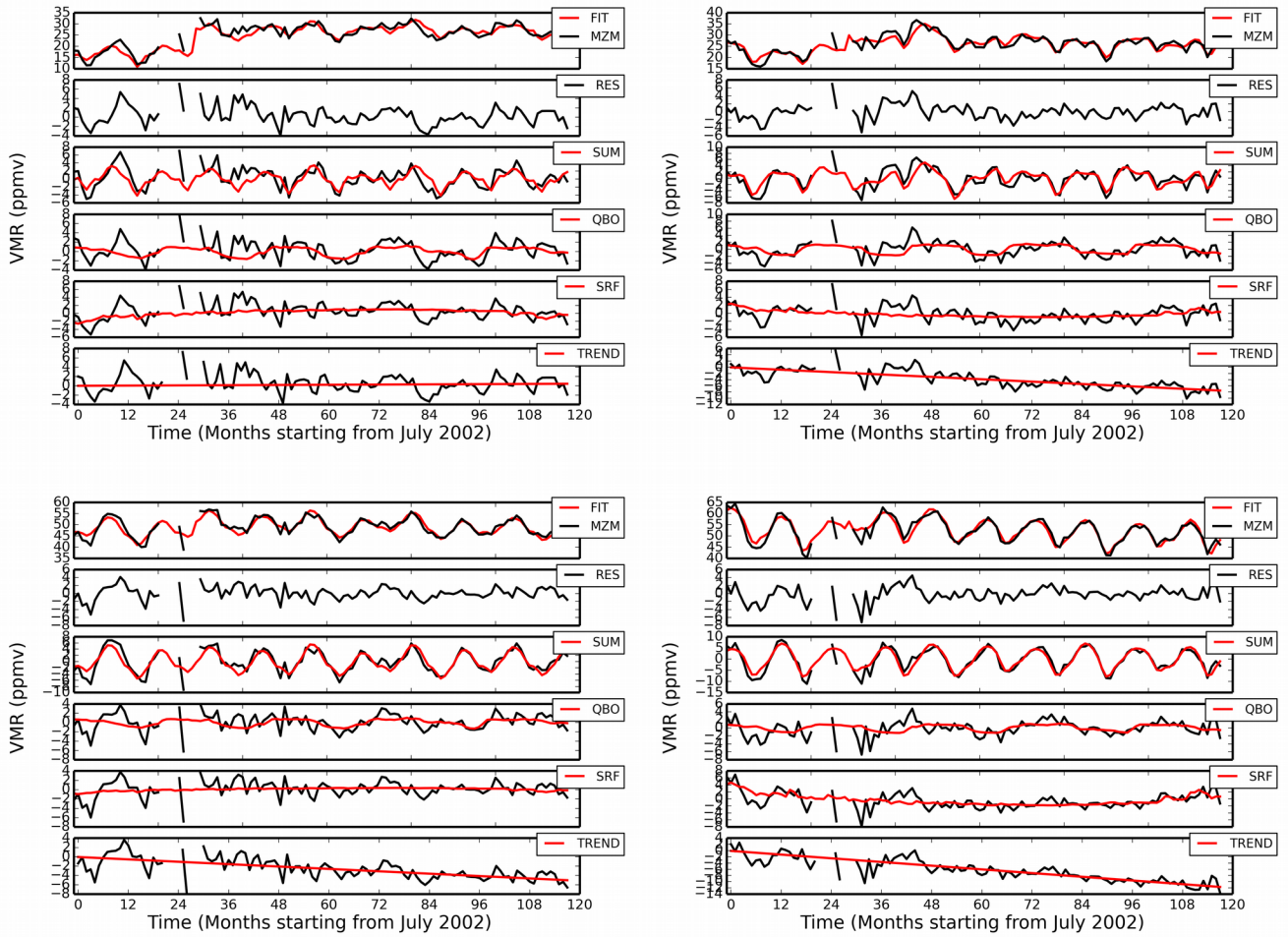


Figure 1A: Contribution of the different terms of the fitting function for 50° - 55° S at 50 hPa (upper left panel), 50° - 55° N at 50 hPa (upper right panel), 50° - 55° S at 80 hPa (bottom left panel) and 50° - 55° N at 80 hPa (bottom right panel). For each panel we report: in the first plot the fitted time series (FIT, red line) and the monthly zonal mean time-series (MZM, black line), in the second plot the residual time-series (RES, black line) calculated as MZM minus FIT; in the third plot the contribution of the sum of the periodicities (SUM, red line) and the MZM minus all the fitted terms excluding SUM (black line); in the fourth plot the contribution of the sum of the two QBO terms (QBO, red line) and the MZM minus all the fitted terms excluding QBO (black line); in the fifth plot the contribution of the solar radio flux (SRF, red line) and the MZM minus all the fitted terms excluding SRF (black line); in the sixth plot the contribution of the trend (TREND, red line) and the MZM minus all the fitted terms excluding TREND (black line).

In Figure 1A we show the contribution of the different terms of the fitting function for different pressure levels and different latitudes (see Fig 1A caption for more details). We can see that the amplitude of the contribution of the different terms of the fitting function depends both on latitude and pressure. In order to avoid discontinuities in the derived

trend values we decided to use the same fitting function (including all terms) for all the pressure / latitude bins, though for some of them, one or more terms of the fitting function may have small or negligible contributions.

c) Comparison with ground stations: is the assumption here that the CCl₄ profile is expected to be constant with altitude all the way through the troposphere? It would have been helpful to show at least a modelled CCl₄ profile to support this. However, the fact that the MIPAS data have a seasonal cycle while the ground station data do not suggests that these must be different air masses, in which case there is presumably also some age difference between the air sampled by MIPAS and the surface air which could explain some of the bias.

A thorough work on modeled CCl₄ has been made by Chipperfield et al. (2016). The modeled CCl₄ profiles shown in that paper are approximately constant in the troposphere. The comparison between CCl₄ retrieved from MIPAS measurements and CCl₄ model data is not a focus in this paper. This comparison will be the subject of a forthcoming work.

To highlight that the comparison is based on the hypothesis of well-mixed troposphere, we added the following sentence at the beginning of Sect. 5.3: “Under the assumption of well-mixed troposphere, we can consider the CCl₄ vertical distribution approximately constant (Chipperfield et al., 2016; Allen et al., 2009)”.

The new reference is:

Chipperfield, M. P., Liang, Q., Rigby, M., Hossaini, R., Montzka, S. A., Dhomse, S., Feng, W., Prinn, R. G., Weiss, R. F., Harth, C. M., Salameh, P. K., Mühle, J., O'Doherty, S., Young, D., Simmonds, P. G., Krummel, P. B., Fraser, P. J., Steele, L. P., Hoppel, J. D., Rhew, R. C., Butler, J., Yvon-Lewis, S. A., Hall, B., Nance, D., Moore, F., Miller, B. R., Elkins, J. W., Harrison, J. J., Boone, C. D., Atlas, E. L., and Mahieu, E.: Model sensitivity studies of the decrease in atmospheric carbon tetrachloride, *Atmos. Chem. Phys.*, 16, 15741-15754, doi:10.5194/acp-16-15741-2016, 2016.

d) Given the data available, it is possible to calculate a *total* atmospheric content of CCl₄, at least the partial column above some pressure surface, and provide the trend of this with time. This would be a much easier quantity for simple

comparison with models or other satellite instruments without having to match details of pressure levels or latitude bands, also for stratospheric chlorine budgets.

We used the approach presented in Sect. 5.1 to estimate also the trend of CCl₄ partial column within two pre-defined pressure levels. For each monthly mean CCl₄ profile referring to a latitude bin we calculated the partial column in the 10 - 100 hPa layer. For each latitude bin we then fitted the time-series of the partial columns using the fitting function (Eq. 1). We finally calculated the weighted average over latitude of the column trends, the weights being the cosine of the average latitude of the bin. For mean hemispheric trends we find $(-8.2 \pm 0.8) \cdot 10^{13} \text{ mol cm}^{-2} \text{ dec}^{-1}$ for SH and $(-12.3 \pm 0.8) \cdot 10^{13} \text{ mol cm}^{-2} \text{ dec}^{-1}$ for NH. Dividing the monthly average columns in each latitude bin by the mission-average column of the same bin we also derive the following relative trends: $(-13.1 \pm 1.7) \% \text{ dec}^{-1}$ for SH and $(-21.7 \pm 1.5) \% \text{ dec}^{-1}$ for NH.

We decided not to include this exercise in the current paper due to the impossibility to make an exhaustive inter-comparison with other measurements. We have found only an atmospheric column trend estimation reported by Rinsland et al. (2012). They measured CCl₄ atmospheric columns over Jungfraujoch (46.5 degN) finding a trend of $(-1.49 \pm 0.08) \cdot 10^{13} \text{ mol cm}^{-2} \text{ yr}^{-1}$. In the 45/50 degN latitudinal band we found a trend of $(-1.15 \pm 0.08) \cdot 10^{13} \text{ mol cm}^{-2} \text{ yr}^{-1}$. As mentioned earlier, the comparison of MIPAS measurements and CCl₄ model data will be the subject of a forthcoming work and we would prefer to include the results of this exercise in that context.

Minor comments

P2 L5: It is not clear from the text whether CCl₄ is an entirely anthropogenic gas or whether there is also some (small?) natural source.

The role of CCl₄ natural sources is not completely clear and the magnitude of these natural emissions is not completely quantified. In the recent SPARC Report (2016) the authors indicate 3-4 Gg/year as the upper limit of the natural emissions.

To highlight this recent result, in the revised paper we added the following sentence in Sect. 1: CCl₄ natural emissions are not completely understood and they are still under discussion. Stratospheric Processes and their Role in Climate (SPARC) community

(SPARC, 2016) recently defined an upper limit of the natural emissions (based on the analysis of old air in firn snow) of 3-4 Gg/year over a total emission estimation of 40 (25-55) Gg/year.

P4 L19: If you mention 'oversampling the limb' you should explain what the size of the field-of-view is.

MIPAS FOV is approximately 3 km in vertical. This information is now included in the mentioned paragraph.

P4 L21: 8 rows for the FR AK, but only 7 for OR.

We rephrased the sentence. This is consistent with the fact that the retrieval grid consists of 8 points (nodes) in the case of FR measurements and of 7 points in the case of OR measurements.

P7 Much of the text here is unnecessary as it is already in the Fig 3 caption.

Here we believe that the information reported in the text is important to understand the details of figure 3 and cannot be delegated uniquely to the figure caption.

P9 Presumably the effect is larger in the antarctic due to the stronger, more stable polar vortex?

OK. We included this comment in the revised paper.

P10 L6: Since the ocean is the major surface sink, and there is more ocean in the southern hemisphere, wouldn't an IHG be expected even in the absence of continued emissions?

If we compare CCl₄ partial lifetime with respect to the ocean sink (209 years (Butler et al., 2016)) with the time needed by an air mass to move from the NH to the SH (around a year), we deduce that, in absence of emissions, the differences between NH and SH concentrations should be negligible. For a more rigorous explanation we refer to Liang et al. 2014.

P11 L5/Fig 6: since Fig 6 is effectively an annual average its difficult to argue which components are persistent and which are seasonal. Perhaps there's an alternative

way of plotting the data to highlight the seasonal differences (eg shift the s. hemisphere data by 6 months before subtracting?)

The figure was built without using a 6-months shift, but we have verified that a shift of 6 months does not change significantly the results since the impact of seasons is reduced by the average over a 7-years period. We revised the text of the paper explaining that the observed differences at high altitudes are not caused by the seasons but they are related to the asymmetry in the magnitude and in the persistence of the subsidence during winter and spring at the two poles.

P11 L14: I can understand why balloon instruments might have better signal/noise than satellite instruments since they can effectively take many scans of the same atmosphere, but I don't understand what is intrinsic to the balloon measurement that gives it high vertical resolution compared to satellites. Indeed the 1.5km spacing of MIPAS-B seems comparable to MIPAS.

We removed this sentence as it was not so important to understand the work presented in Section 4.1. The original intention was to explain that with a given angular aperture of the instrument FOV, the vertical resolution achieved from a stratospheric balloon platform is finer than that achieved from the satellite because the balloon is much closer to the sampled atmosphere. However MIPAS-B and MIPAS/ENVISAT instruments do not have the same angular FOV aperture.

P15 L12: Given that CCl₄ is a relatively long-lived gas with no diurnal variation, and that both MIPAS and ACE-FTS obtain relatively uniform sampling in longitude, I wonder why you didn't simply compare zonal means of both datasets (interpolating MIPAS to the appropriate latitude for ACE-FTS each day) rather than look for profile-by-profile coincidences which could contain a latitude bias or end up just selecting MIPAS ascending or descending node observations (with the associated GRAD error).

As highlighted in the plot in the bottom panel of Fig. 3, in this part of the mission the GRAD error is expected to show a maximum value of only 3% at 15 km (approximately 120 hPa) and to rapidly decrease at higher altitudes. For this reason the GRAD error is not expected to play an important role in the inter-comparison with ACE. Moreover, since the horizontal resolution of MIPAS is at least as broad as 300 km for the weakest

species (see von Clarmann, T., De Clercq, C., Ridolfi, M., Höpfner, M., and Lambert, J.-C.: The horizontal resolution of MIPAS, Atmos. Meas. Tech., 2, 47-54, doi:10.5194/amt-2-47-2009, 2009) the matching criterion we use (300 km and 3 hrs) is quite stringent. Note that with our used matching method we also avoid the interpolation error that would be implied by the approach suggested by the reviewer.

P15 L15: Again much of the text repeats what is in the figure caption, although it takes a while before explaining what I really wanted to know, which is the distinction between 'standard deviation of the mean' and 'standard deviation of the differences'. The former is just the latter divided by root(N), is that right?

Yes, right. We modified the text to include this detail.

P17 Eq(1): I agree with the approach but the term 'offset parameters' confused me - offset relative to what? Perhaps just 'constant parameters'.

Done. We have replaced “offset parameters” with “ constant parameters”.

Typographic/grammatical comments

P1 L1: no need for capital C in 'Carbon tetrachloride'

Done.

P1 L12: 20-50 rather than 20/50 if this indicates a range of latitudes rather than a particular pair of latitudes

This sentence has been deleted.

P3 L9: Similarly.

Done.

P2 L33: Suggest 'limits' rather than 'edges'.

Done.

P3 L14: 'where' rather than 'were'

Done.

P15 L6: Suggest 'extends' rather than 'goes'

Done.

Fig 5: some vertical lines at the year boundaries would be helpful.

Done. We have modified Fig. 5 adding vertical dashed lines at the year boundaries. This information is now reported also in the caption.

Fig 6: 'degN' for the latitude axis should presumably just be 'deg' here.

Done.